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Very Fast Inorganic Crystal Scintillators

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ABSTRACT

Future HEP experiments at the energy and intensity frontiers require fast inorganic crystal scintillators with excellent radiation hardness to face the challenges of unprecedented event rate and severe radiation environment. This paper reports recent progress in application of very fast inorganic scintillators in future HEP experiments, such as thin layer of LYSO crystals for a shashlik sampling calorimeter and a precision TOF detector proposed for the CMS upgrade at the HL-LHC, undoped CsI crystals for the Mu2e experiment at Fermilab and yttrium doped BaF₂ crystals for the proposed Mu2e-II experiment. Applications for Gigahertz hard X-ray imaging will also be discussed.

Keywords: Crystal, scintillator, calorimeter, radiation damage, fast timing

1. INTRODUCTION

Fast and radiation hard inorganic crystal scintillators are needed for future HEP experiments at the energy and intensity frontiers. For experiments to be operated at the HL-LHC with 3,000 fb⁻¹, for example, crystals should survive an environment with absorbed dose of 100 Mrad, charged hadron fluence of 6×10^{14} cm⁻² and fast neutron fluence of 3×10^{15} cm⁻². For future HEP experiments at the intensity frontier, such as Mu2e-II, ultra-fast crystals are needed to face the challenge of unprecedented event rate as well as severe radiation.

Table 1 Inorganic crystals commonly used in HEP experiments

Crystal	NaI(Tl)	CsI(Tl)	CsI	BaF ₂	BGO	LYSO(Ce)	PWO	PbF ₂
Density (g/cm ³)	3.67	4.51	4.51	4.89	7.13	7.40	8.3	7.77
Melting Point (°C)	651	621	621	1280	1050	2050	1123	824
Radiation Length (cm)	2.59	1.86	1.86	2.03	1.12	1.14	0.89	0.93
Molière Radius (cm)	4.13	3.57	3.57	3.10	2.23	2.07	2.00	2.21
Interaction Length (cm)	42.9	39.3	39.3	30.7	22.8	20.9	20.7	21.0
Refractive Index ^a	1.85	1.79	1.95	1.50	2.15	1.82	2.20	1.82
Hygroscopicity	Yes	Slight	Slight	No	No	No	No	No
Luminescence ^b (nm) (at peak)	410	550	310	300 220	480	402	425 420	?
Decay Time ^b (ns)	245	1220	26	650 0.9	300	40	30 10	?
Light Yield ^{b,c} (%)	100	165	4.7	36 4.1	21	85	0.3 0.1	?
d(LY)/dT ^b (%/°C)	-0.2	0.4	-1.4	-1.9 0.1	-0.9	-0.2	-2.5	?
Experiment	Crystal Ball	BaBar BELLE BES-III	KTev S.BELLE Mu2e-I	(GEM) TAPS Mu2e-II?	L3 BELLE HHCAL?	COMET & CMS (Mu2e & SperB)	CMS ALICE PANDA	A4 g-2 HHCAL?

a. at peak of emission; b. up/low row: slow/fast component; c. QE of readout device taken out.

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Table 1 lists basic properties of crystals commonly used to build crystal calorimeters for HEP experiments, where calorimeters with experiment names in brackets were proposed but not constructed. Among the crystals listed in Table 1, bright, fast and radiation hard lutetium yttrium oxyorthosilicate ($\text{Lu}_{2(1-x)}\text{Y}_x\text{SiO}_5\text{:Ce}$ or LYSO) crystals were proposed for a LYSO/W/Quartz capillary sampling calorimeter [1] and a precision time of flight (TOF) layer for the CMS upgrade for the HL-LHC [2]. LYSO crystals are also proposed to construct a total absorption calorimeter for the COMET experiment at KEK [3], and a 3D calorimeter for the proposed HERD experiment in space [4]. Undoped CsI crystals are used to construct a calorimeter for the Mu2e experiment at Fermilab [5]. BaF_2 crystals have a unique fast scintillation light with sub-ns decay time, but also a slow scintillation light component with 600 ns decay time. Recently developed yttrium doped BaF_2 crystals with effective slow component suppression have a great potential for an extra fast calorimeter for Mu2e-II [6]. They may also be used in the proposed MaRIE facility at Los Alamos [7], where unprecedented fast X-ray imaging requires ultra-fast detector response for hard X-rays [8].

2. PULSE SHAPE AND RISE TIME OF FAST SCINTILLATORS

Crystal time resolution depends on the signal to noise ratio for the rise time measurement. While the intrinsic rising time of most crystals is as fast as a few tens ps [9], Fig. 1 shows the rise time measured for ten crystal samples of $1.5X_0$ size by using a Hamamatsu R2059 PMT and a fast digital scope. The fast rise time of about 1.5 and 1.6 ns observed respectively for BaF_2 and LYSO is limited by the PMT rise time 1.3 ns at 2,500 V bias and the rise time of 0.14 ns of the Agilent MSO9254A (2.5 GHz) DSO. The measured rise time is also faster for the same crystal with a black wrapping, where the light propagation in crystal is minimized [10].

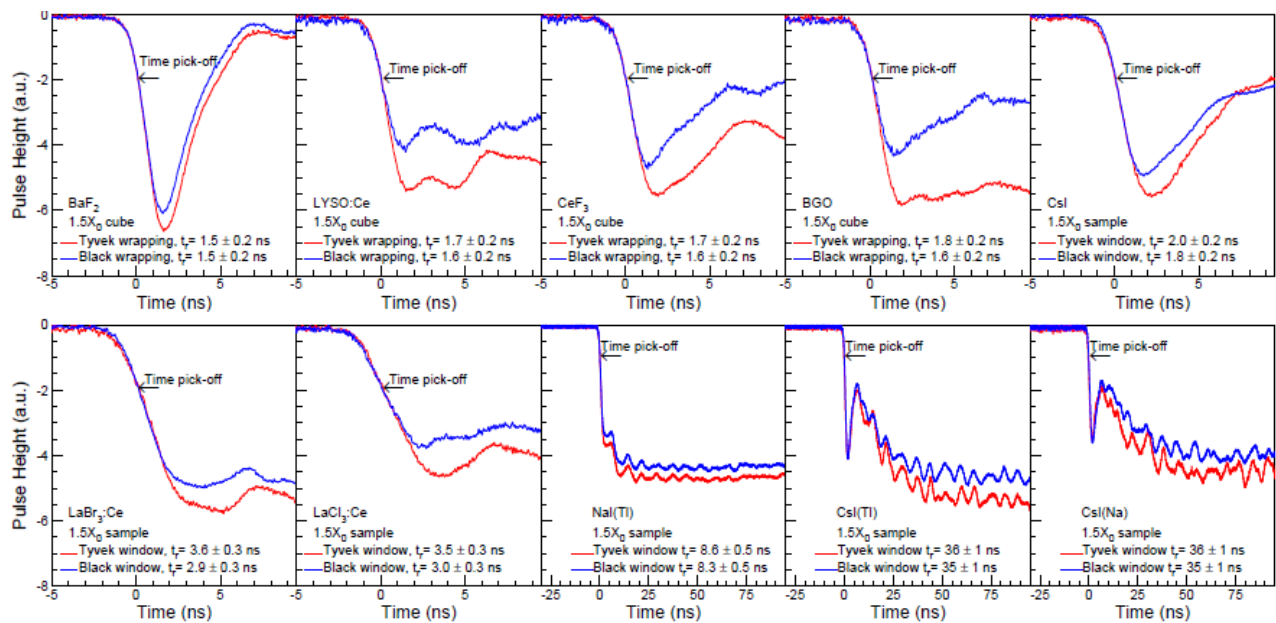


Fig. 1. Scintillation rising time measured for ten crystal samples of $1.5 X_0$

Table 2 lists the figure of merit values on time resolution for various crystal detectors, which is defined as the light output in the 1st, or the 1st 0.1, ns [10]. It is clear that the best crystal scintillators for ultra-fast timing are BaF_2 , LSO:Ca,Ce and LYSO:Ce. LaBr_3 is a material with high potential theoretically, but suffers from scattering centers in the crystal as well as its intrinsic hygroscopicity. On the other hand, the decay time of most fast scintillators at a level of few tens ns would cause pile-up for calorimeters designed for very fast rate capability and GHz x-ray imaging with a x-ray frame rate of a few ns. It thus is crucial to develop very fast inorganic scintillator capable to provide clean signal pulse of a few ns width. Among all scintillators shown in Fig. 1, the BaF_2 crystals are the only one with this capability provided that its slow scintillation component is suppressed.

Table 2. Figure of Merit for Time Resolution for Various Crystal Scintillators

Crystal Scintillators	Relative LY (%)	A ₁ (%)	τ ₁ (ns)	A ₂ (%)	τ ₂ (ns)	Total LO (p.e./MeV, XP2254B)	LO in 1ns (p.e./MeV, XP2254B)	LO in 0.1ns (p.e./MeV, XP2254B)	LY in 0.1ns (photons/MeV)
BaF ₂	40.1	91	650	9	0.9	1149	71.0	11.0	136.6
LSO:Ca,Ce	94	100	30			2400	78.7	8.0	110.9
LSO/LYSO:Ce	85	100	40			2180	53.8	5.4	75.3
CeF ₃	7.3	100	30			208	6.8	0.7	8.6
BGO	21	100	300			350	1.2	0.1	2.5
PWO	0.377	80	30	20	10	9.2	0.42	0.04	0.4
LaBr ₃ :Ce	130	100	20			3810	185.8	19.0	229.9
LaCl ₃ :Ce	55	24	570	76	24	1570	49.36	5.03	62.5
NaI:Tl	100	100	245			2604	10.6	1.1	14.5
CsI	4.7	77	30	23	6	131	7.9	0.8	10.6
CsI:Tl	165	100	1220			2093	1.7	0.2	4.8
CsI:Na	88	100	690			2274	3.3	0.3	4.5

3. QUALITY OF PREPRODUCTION CSI CRYSTALS FOR MU2E

The Mu2e experiment at Fermilab is constructing a fast calorimeter using undoped CsI crystals. 72 preproduction CsI crystals from three vendors were characterized. Fig. 2 compares measured data with the Mu2e specifications (red dashed lines), showing that most crystals meet the specifications on light output, FWHM energy resolution, light response uniformity, F/T (Fast/Total) ratio, γ -ray induced noise and radiation hardness. Some crystals failed specifications on F/T ratio and γ -ray induced noise because of significant slow scintillation component. Excellent correlations have been observed between the light output and the energy resolution, and the dark current, the γ -ray induced readout noise and the F/T ratio, confirming that they are of the same origin [11].

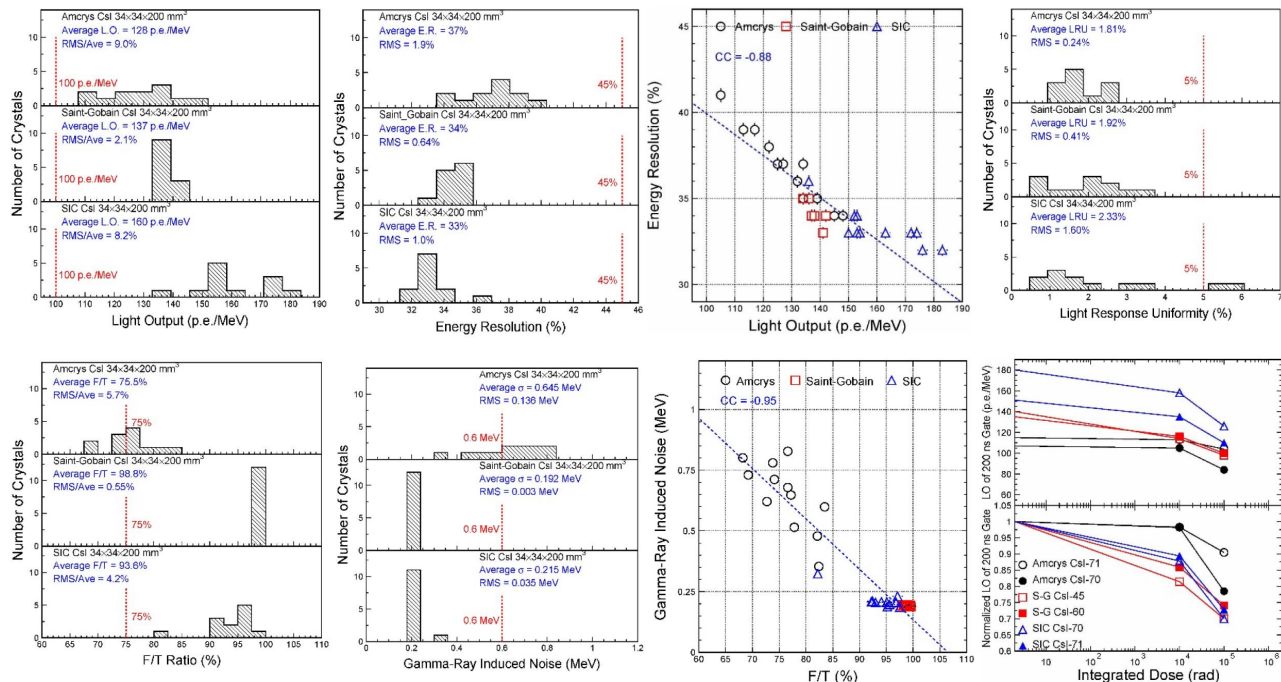


Fig. 2. Quality of preproduction undoped CsI crystals of 3.4 x 3.4 x 20 cm

4. YTTRIUM DOPED BaF₂ CRYSTALS

BaF₂ crystal has a very fast scintillation component peaked at 220 nm with sub-ns decay time, which provides a solid foundation for a very fast calorimetry. It, however, has also a slow scintillation component peaked at 300 nm with 600 ns decay time and a five times brightness of its fast component, which would cause pileup in an environment of fast rate. Two approaches are used to reduce the pileup caused by slow component: selective doping with rare earth (La, Y, and Ce) in BaF₂ [12] and selective readout with solar blind photodetector [13]. The top left photo of Fig. 3 shows a set of nine yttrium doped BaF₂ crystal samples of $\Phi 18 \times 21$ mm doped with yttrium from 0 to 7 mol% from Beijing Glass Research Institute (BGRI). The top middle and right plot shows their x-ray excited luminescence (XEL) and transmittance spectra respectively. The bottom left plot shows their light output as a function of integration time. The bottom middle and right plots show their light output in 50 and 2,500 ns gates and the corresponding ratios respectively as a function of the level of yttrium doping in mol%. It was found that the optimized yttrium doping in BaF₂ is about 5mol%, which increases the F/S (Fast/Slow) ratio from 1/5 to 5/1 while the amount of fast light is unchanged. The crystals of this nature is expected to find a broad application in future HEP experiments and GHz x-ray imaging.

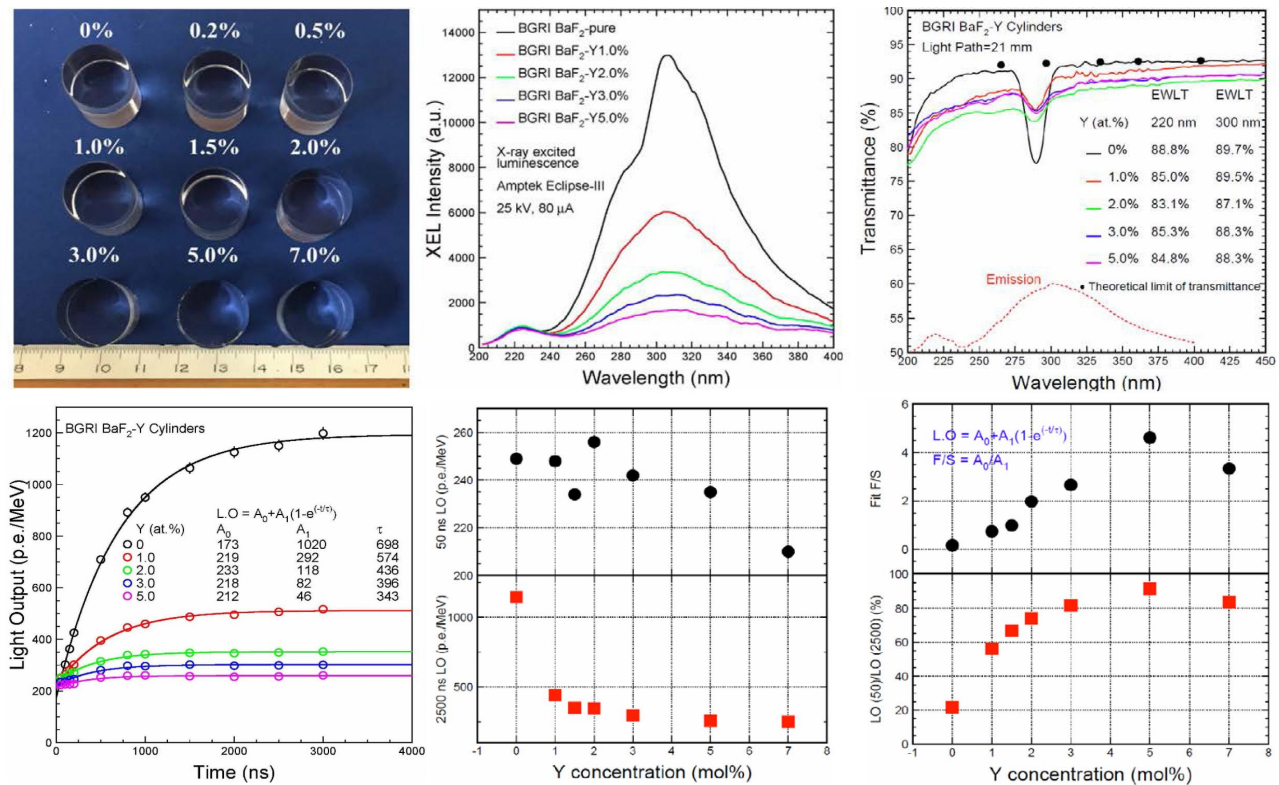


Fig. 3. Performance of Yttrium doped BaF₂ Crystals of $\Phi 18 \times 21$ mm

5. NEUTRON INDUCED RADIATION DAMAGE IN FAST CRYSTALS

One crucial issue for applications of scintillation crystals in future HEP experiments is radiation damage in severe radiation environment, such as the HL-LHC. While radiation damage induced by ionization dose in inorganic crystal scintillators is well understood, investigations are on-going to understand radiation damage caused by hadrons, including both charged hadrons and neutrons. Two neutron irradiation experiments were carried out in 2015 (Exp. 6991) and 2016 (Exp. 7332) in the East Port of the Target-4 area (top left plot in Fig. 4), about 1.2 m away from the neutron production target at the Weapons Neutron Research facility of the Los Alamos Neutron Science Center (WNR of LANSCE). The top middle plot of Fig. 4 shows the particle rate as a function of energy for neutrons, photons and protons, tallied on the

largest sample volume (averaging) around the samples, which are calculated by using the Monte Carlo N-Particle eXtended (MCNPX) transport package developed by LANL. In the 2016 run, 36 samples of 5 mm thick were irradiated for 21.2, 46.3 and 120 days in three groups of 12 each (top right photo of Fig. 4), where half samples in each group were shielded by 5 mm Pb, and other half not. The corresponding fast neutron ($>1\text{MeV}$) fluence and ionization dose were $3.94, 8.58$ and $19.3 \times 10^{14} \text{ n/cm}^2$ and 42/30, 52/65 and 207/147 Mrad respectively for three groups without/with lead shielding.

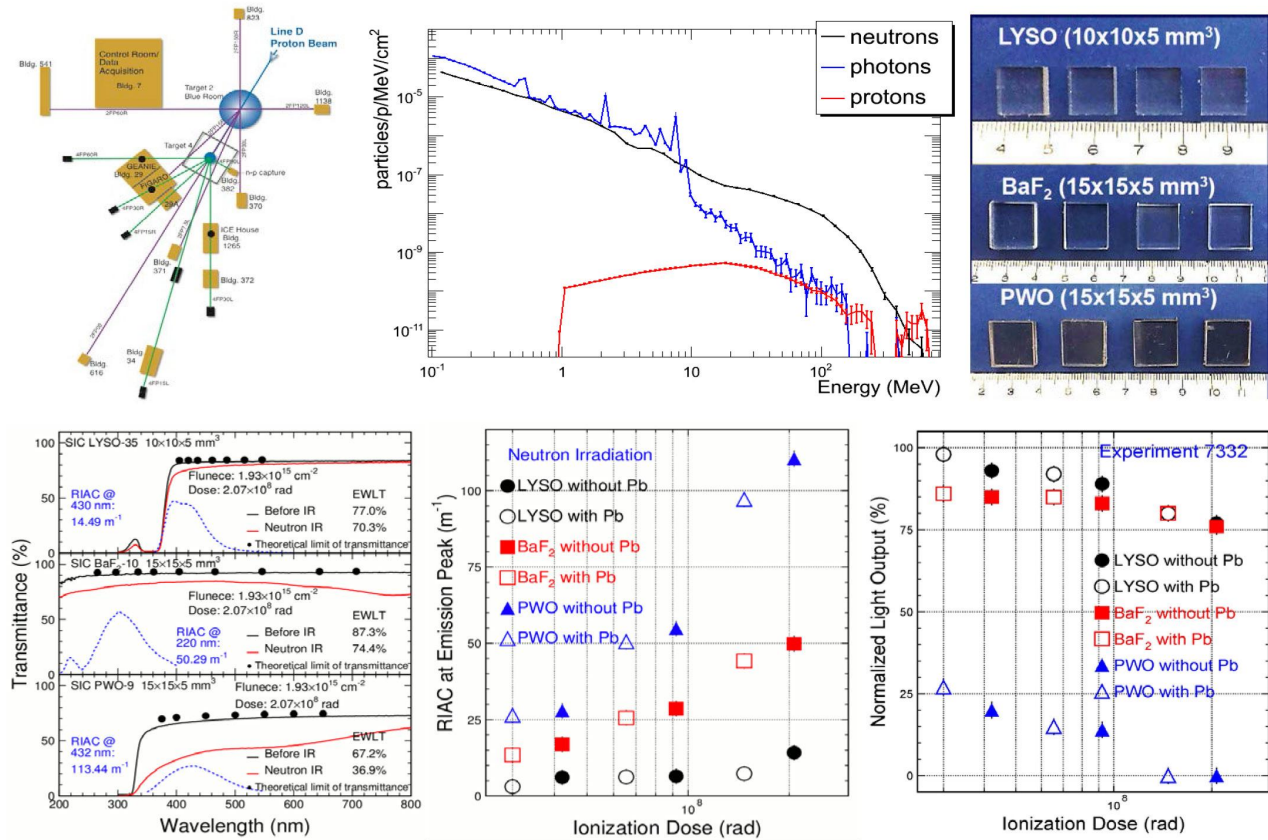


Fig. 4. Neutron Irradiation Experiment for LYSO, BaF₂ and PWO crystals at LANSCE

The bottom left plot of Fig. 4 shows transmittance spectra before and after irradiation for one sample each of LYSO (top), BaF₂ (middle) and PWO (bottom) without lead shielding in the group 3 of the 2016 run. Also listed in the figure is the numerical values of the emission weighted longitudinal transmittance (EWLT) and the radiation induced absorption coefficient (RIAC) after the irradiation. It is clear that LYSO and BaF₂ are much more radiation hard than PWO. The bottom middle and right plots of Fig. 4 present the RIAC and the normalized light output (LO) as a function of the ionization dose, showing that the contribution of fast neutron induced damage, if any, is much less than that from the ionization dose. This observation confirms an early observation of no neutron specific damage in PWO crystals [14], and indicates that neutrons are very different from charged hadrons in hadron induced radiation damage in scintillating crystals.

6. SUMMARY

LYSO is a robust scintillators against ionization dose as well as charged and neutral hadrons expected at the HL-LHC. Commercially available undoped CsI crystals satisfy the Mu2e requirements. Commercially available undoped BaF₂ crystals provide sufficient fast light with sub-ns decay time and excellent radiation hardness beyond 100 Mrad and

$1 \times 10^{15} \text{ p cm}^{-2}$. They promise a very fast and robust calorimeter in a severe radiation environment. Yttrium doping in BaF₂ crystals increases the F/S ratio from 1/5 to 5/1 without using a selected readout, while maintaining the amount of fast scintillation light unchanged. The slow contamination at this level is already much less than the commercially available undoped CsI crystals. This novel crystal promises a very fast calorimetry for future HEP experiments and an effective frontend detector for GHz x-ray imaging. Results of the experiments 6991 and 7332 at LANSCE show fast neutrons up to $2 \times 10^{15} \text{ n/cm}^2$ do not damage LYSO, BaF₂ and PWO crystals. Our plan is to further investigate radiation hardness of Y:BaF₂ crystals, and pay special attention to photodetector with DUV response: LAPPD, Si or diamond based solid state detector [15].

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